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Effective conductivity for interface scattering in metal-matrix composites

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The contribution to the resistivity of metal-matrix composites resulting from electrons scattering from the surface of the embedded phase has long been recognized, but quantitative assessments of this effect are practically unknown. We present a theory that can serve as a starting point for such investigations. The theory focuses on an effective local conductivity for scattering, which rises steadily from a minimum at the scattering surface to the matrix value several mean-free-path lengths into the bulk. By making suitable assumptions, we are able to give a closed-form expression for this local conductivity, which is insensitive to the geometry of the scattering surface. Our analytical results compare favorably with recent numerical calculations for the scattering of electrons from the external surface of a long cylinder.

I. INTRODUCTION

In the theory of composite we attempt to assign to a multiple-phase (heterogeneous) material an effective constant, such as the conductivity, given the properties of the component (homogeneous) phases, and their morphology. Since full knowledge of structural details is rarely available (or even desirable), a variety of models has emerged which differ according to the structural approximations made. Nonetheless, a few features are common to all, and can be established under very general assumptions. Among them we find that the effective conductivity of a composite is a homogeneous function of degree one of the component conductivities.[1] For metal-matrix composites formed with nonconducting fibers, this implies that the effective longitudinal and transverse conductivities are both linear functions of the matrix conductivity and, hence, are linear to each other. In particular, the matrix conductivity, the longitudinal conductivity, and the transverse conductivity should all obey the same temperature law. However, conductivity *data* do not always conform to this expectation,[2] and the temperature law itself may be unfamiliar.

Discrepancies typically appear in composites with a high fiber content and at temperatures well below room temperature. They can be explained qualitatively by postulating an additional contribution to the sample resistivity due to scattering at the fiber-matrix interface. This scattering results in a local conductivity $\vec{\sigma}(\mathbf{r})$, which is smallest at the interface and steadily approaches the bulk value $\vec{\sigma}_0$ for the matrix far removed from the fiber surface. The characterization of interface scattering in terms of an effective local conductivity is well suited to composites; indeed, if the form of $\vec{\sigma}(\mathbf{r})$ were known, the effects of interface scattering could be brought within the fold of traditional composite theories. In this paper we will show that a closed-form expression for $\vec{\sigma}(\mathbf{r})$ can be found under certain simplifying assumptions, and that this form is insensitive to the precise geometry of the scattering surface.

Our goals are similar to those addressed in the classic work by Dingle,[3] who studied the effects of surface scattering on the resistivity of thin wires and films. The extension of Dingle's work to electron scattering from the *external* surface of a cylinder (the fiber-matrix interface of metal-matrix composites) was made recently by Roig and Schoutens,[4] although their low-temperature results are suspect.[5] Both of these efforts, however, were predominantly numerical computations carried out in special geometries, and did not emphasize the local-conductivity concept so important for general composite applications.

II. THE LOCAL CONDUCTIVITY FOR INTERFACE SCATTERING

The conductivity of metals typically is studied in the (semiclassical) context of the Boltzmann transport equation, and the so-called *relaxation-time approximation.* This is the approach adopted here, although it must be admitted that the concept of a relaxation time may be called into question when the mean free path for scattering becomes large, as it does in pure specimens at low temperatures. In the semiclassical view, the phase-space distribution of carriers [electrons] is described by a *distribution function* $f(\mathbf{r},\mathbf{v})$, taken as the sum of an equilibrium term f^0 and an "out-of-balance" component $g(\mathbf{r},\mathbf{v})$ arising from the application of a steady electric field $\mathbf{E}$. Since f^0 is spatially uniform in bulk specimens (indeed, proportional to the Fermi distribution), the out-of-balance component $g(\mathbf{r},\mathbf{v})$ must satisfy

$$\mathbf{v}_k\cdot\nabla g+\frac{g(\mathbf{r},\mathbf{v}_k)}{\tau}=\left[-\frac{\partial f^0}{\partial\varepsilon}\right]\mathbf{v}_k\cdot e\mathbf{E}\ . \tag{1}$$

Here τ denotes the relaxation time for everything but surface scattering (e.g., impurities, imperfections, thermal vibrations), and $\mathbf{v}_k$ is the velocity of carriers with wave vector $\mathbf{k}$ and energy $\varepsilon(\mathbf{k})$, as befits conduction electrons in a metal. The steady current density $\mathbf{J}(\mathbf{r})$ is calculated from $g(\mathbf{r},\mathbf{v}_k)$ as

$$\mathbf{J}(\mathbf{r})=\int e\mathbf{v}_k g(\mathbf{r},\mathbf{v}_k)d^3k\ . \tag{2}$$

Since $g(\mathbf{r},\mathbf{v}_k)$ will be proportional to the applied field, the sample conductivity follows immediately from Eq. (2) once the proper solution to Eq. (1) is found. Note that the emergence of a local spatially varying conductivity implies that $g(\mathbf{r},\mathbf{v}_k)$ will vary in a nontrivial way with $\mathbf{r}$ in the neighborhood of the fiber surface.

The general solution to Eq. (1) expressed in a form convenient for our purposes is afforded by Chamber's formula[6]

$$g(\mathbf{r},\mathbf{v}_k)=\left[-\frac{\partial f^0}{\partial\varepsilon}\right]e\mathbf{E}(\mathbf{r})\cdot\boldsymbol{\lambda}_k \times[1+G(\mathbf{v}_k,\mathbf{r}_B)e^{-|\mathbf{r}-\mathbf{r}_B|/\lambda_k}]\ , \tag{3}$$

where we have written $\boldsymbol{\lambda}_k=\tau\mathbf{v}_k$ for the vector mean free path of the electron. $\mathbf{r}_B$ is that point on the bounding (fiber) surface derived from $\mathbf{r}$ by extrapolation along the direction of $\mathbf{v}_k$ (see Fig. 1), and $G(\mathbf{v}_k,\mathbf{r}_B)$ is a (presumed known) function accounting for the condition and geometry of the fiber surface. The second term in Eq. (3) contains the correction to the conductivity arising from interface scattering. Indeed, $|\mathbf{r}-\mathbf{r}_B|\gg\lambda_k$ far from the interface, leaving the usual solution for the bulk material. For surfaces rough on the scale of the carrier wavelength (Fermi wavelength), we take $G=-1$, independent of $\mathbf{v}_k$ and $\mathbf{r}_B$. This corresponds to *diffuse* scattering, and is the choice adopted here for simplicity. Other choices for $G(\mathbf{v}_k,\mathbf{r}_B)$ are computationally feasible only if the surface geometry is sufficiently simple;[7] in keeping with our objectives, these will not be considered further.

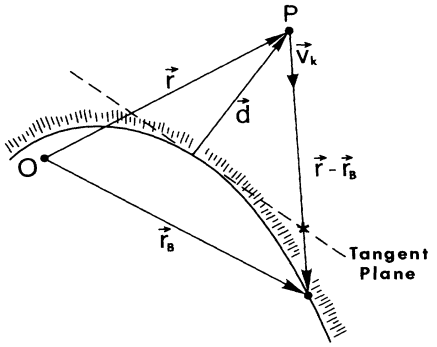

FIG. 1. Construction showing the relationship between $\mathbf{r}$, $\mathbf{r}_B$, and $\mathbf{v}_k$. If the surface directly below P is locally flat on the scale of λ, $|\mathbf{r}-\mathbf{r}_B|$ can be approximated by the distance from P to the tangent plane.

The structure of Eq. (3) for $g(\mathbf{r},\mathbf{v}_k)$ when substituted into Eq. (2) leads to a local conductivity (tensor) of the form

$$\overleftrightarrow{\sigma}(\mathbf{r})=\overleftrightarrow{\sigma}_0+\Delta\overleftrightarrow{\sigma}(\mathbf{r})\ , \tag{4}$$

where $\Delta\overleftrightarrow{\sigma}$, the deviation from the bulk value $\overleftrightarrow{\sigma}_0$, is given by

$$\Delta\overleftrightarrow{\sigma}(\mathbf{r})=-\frac{e^2}{4\pi^3\hbar}\int_{\mathrm{FS}}dS_F\mathbf{n}_k\mathbf{n}_k\lambda_k e^{-|\mathbf{r}-\mathbf{r}_B|/\lambda_k}\ . \tag{5}$$

The integral in Eq. (5) is taken over the Fermi surface (FS) of the host metal, and $\mathbf{n}_k$ denotes the unit vector in the direction $\mathbf{v}_k$. Our treatment of this integral is based on the following observations: (i) $\Delta\overleftrightarrow{\sigma}(\mathbf{r})$ is insignificant unless the field point $\mathbf{r}$ lies within a few mean free paths λ_k of the fiber surface, and (ii) sufficiently close to the fiber, the surface appears nearly flat; if $\boldsymbol{\lambda}$ sets the distance scale for proximity to the surface, then local flatness is assured if $R\gg\lambda$, where R is the radius of curvature at any point on this surface. Surrounding the fiber, then, we envision a sheath of thickness $\sim\lambda$ within which $\Delta\overleftrightarrow{\sigma}$ is appreciable; our computational method assumes the fiber surface appears nearly flat when viewed from anywhere in this sheath. That being so, we can approximate $|\mathbf{r}-\mathbf{r}_B|$ as the distance along the direction $\mathbf{v}_k$ from $\mathbf{r}$ to the tangent plane defined by the surface normal passing through the field point P (see Fig. 1), or

$$|\mathbf{r}-\mathbf{r}_B|\approx-\frac{d(\mathbf{r})}{\cos(\mathbf{d}\cdot\mathbf{v}_k)}\ . \tag{6}$$

Here $\mathbf{d}(\mathbf{r})$ is a vector normal to the scattering surface and drawn *inward toward the bulk,* as shown in the figure.

To complete the evaluation of $\Delta\overleftrightarrow{\sigma}$, we assume the same mean free path $\boldsymbol{\lambda}$ for all electrons at the Fermi surface of the host metal, and approximate this Fermi surface with a sphere. Then local axes at P parallel and perpendicular to $\mathbf{d}$ become principle axes for $\Delta\overleftrightarrow{\sigma}$. Taking the polar axis of spherical coordinates in k-space along $\mathbf{d}$ [so that $\cos(\mathbf{d}\cdot\mathbf{v}_k)=\cos\theta$], and introducing $z=|\mathbf{d}(\mathbf{r})|/\lambda$, we find in the direction of $\mathbf{d}$ (which is actually normal to the surface)

$$\begin{aligned}\Delta\sigma_\perp(\mathbf{r})&=-\tfrac{3}{2}\sigma_0\int_{\pi/2}^{\pi}\sin\theta\cos^2\theta e^{z/\cos\theta}d\theta\\&=-\tfrac{3}{2}\sigma_0\int_1^\infty e^{-zu}\frac{du}{u^4}\\&=-\frac{3}{2}\sigma_0E_4(z)\ .\end{aligned} \tag{7}$$

Here we have identified

$$\sigma_0=\frac{e^2\lambda k_F^2}{3\pi^2\hbar} \tag{8}$$

as the (scalar) conductivity of the bulk metal in the same approximation, and $E_4(z)$ as one of a class of exponential integral functions.[8] In the same way, we find for the conductivity change tangential to the interface

$$\Delta\sigma_{\parallel}(\mathbf{r}) = -\frac{3}{8\pi}\sigma_0\int_{\pi/2}^{\pi}\sin^3\theta e^{z/\cos\theta}d\theta$$
$$= -\frac{3}{8\pi}\sigma_0\int_1^{\infty}e^{-zu}\left[1-\frac{1}{u^2}\right]\frac{du}{u^2}$$
$$= -\frac{3}{8\pi}\sigma_0[E_2(z)-E_4(z)] \ . \quad (9)$$

Notice that the integrals in Eqs. (7) and (9) are restricted to polar angles $\theta > \frac{1}{2}\pi$; for smaller angles $|\mathbf{r}-\mathbf{r}_B|$ is effectively infinite, since electrons moving in these directions never reach the surface.

III. DISCUSSION

The expression of $\Delta\vec{\sigma}(\mathbf{r})$ in terms of exponential integral functions fulfills our prime objective laid down in Sec. I. Not surprisingly, the natural variable for this problem has turned out to be $z=|\mathbf{d}(\mathbf{r})|/\lambda$. Deviation from the bulk conductivity is greatest at the fiber surface ($z=0$) where $E_n(0)=(n-1)^{-1}$ leads to the limits

$$\frac{|\Delta\sigma_{\perp}|}{\sigma_0}\to\frac{1}{2} \ , \quad \frac{|\Delta\sigma_{\parallel}|}{\sigma_0}\to\frac{1}{4\pi}\simeq 0.08 \ . \quad (10)$$

The changes are not substantial, especially for the tangential component where the deviation is less than 10% of the bulk value. From the asymptotic forms

$$E_n(z)\sim\frac{e^{-z}}{z}\left[1-\frac{n}{z}+O\left[\frac{1}{z^2}\right]\right] , \quad (11)$$

we see that $\vec{\sigma}(\mathbf{r})$ approaches $\vec{\sigma}_0$ in essentially exponential fashion with characteristic length λ, the rise being somewhat sharper for the component parallel to the fiber surface.

As a check on these results, consider the geometry of Ref. 4 wherein a single, infinitely long and perfectly straight nonconducting fiber is embedded in a metallic host. Electric conduction takes place along the fiber axis, driven by a uniform field $\mathbf{E}$. Yet on account of interface scattering, $\mathbf{J}(\mathbf{r})$ is nonuniform and the *apparent* conductivity σ of the specimen becomes

$$\sigma = \frac{1}{|\mathbf{E}|A}\int_{A_c}J_{\parallel}(\mathbf{r})dA = \frac{1}{A}\int_{A_c}\sigma_{\parallel}(\mathbf{r})dA \ . \quad (12)$$

The integrals in Eq. (12) are taken over the cross section A_c for the region of interest, and A is the area of that cross section. In polar coordinates (r,θ) centered on a fiber with radius a, we see that $|\mathbf{d}(\mathbf{r})|=r-a$ and Eq. (12) becomes

$$\frac{\sigma}{\sigma_0} = 1-\frac{3}{8\pi A}\int_0^{z_c}2\pi(\lambda z+a)[E_2(z)-E_4(z)]\lambda dz$$
$$\approx 1-\frac{2\pi a^2}{A}I(k) \ . \quad (13)$$

In writing the second line, we have extended the upper limit of integration to infinity (with negligible error) and introduced the function

$$I(k) = \frac{3}{2\pi}\int_0^{\infty}\left[\frac{1}{2k}+\frac{z}{k^2}\right][E_2(z)-E_4(z)]dz$$
$$= \frac{3}{16\pi}\left[\frac{1}{k}+\frac{16}{15}\frac{1}{k^2}\right] \quad \left[k=\frac{2a}{\lambda}\right] \quad (14)$$

to facilitate comparison with Ref. 4. There, $I(k)$ was found from numerical evaluation of a triple integral. The results, represented by a straight line on a log-log plot, seem to conform to the power law

$$I(k)\simeq 0.27\frac{1}{k^{1.08}} \ . \quad (15)$$

This functional dependence is in close agreement with our predicted form Eq. (14), through the latter gives $I(1)=31/80\pi$ (0.123) as compared to the numerical result 0.27. The discrepance is not at all surprising in view of (1) the numerical problems alluded to in Ref. 4 in handling the triple integral, and (2), the criterion for local flatness, which in the present context requires $k\gg 1$ in Eq. (14) in order to obtain reliable values. Thus it seems safe to conclude that we are able to recover the essential results of Ref. 4, but with a good deal less effort.

Of course, the full power of our formulation becomes apparent only in the complex geometry of real composites, where field inhomogeneities (due to fiber arrangement, local charging effects, etc.) are prevalent. This problem will be explored further in a subsequent publication. We close this paper with the following general remarks and observations:

(i) The results of Eqs. (7) and (9) for $\vec{\sigma}(\mathbf{r})$ may have greater validity than expected, i.e., even if the interface is not locally flat on the scale of λ. Certainly this is suggested by the success of Eq. (14) with $k\sim 1$ in describing scattering from the surface of a long cylinder. More generally, we see that points $\mathbf{r}_B$ on the surface directly below $\mathbf{r}$ contribute most to the integral in Eq. (5), and our approach always handles those contributions correctly.

(ii) The tensor character of $\vec{\sigma}(\mathbf{r})$ complicates the theory, but is indispensible in some situations. Certainly if the fiber were conducting, there could be current flow normal as well as tangential to the fiber surface. Even nonconducting fibers crowded together in irregular arrangements could easily distort any applied field enough to produce local fields with large normal components near (but not at) the fiber surface. To retain the simplicity of a scalar description in such cases, we might consider replacing the conductivity tensor $\vec{\sigma}(\mathbf{r})$ by its directional average

$$\bar{\sigma}(\mathbf{r}) = \tfrac{2}{3}\sigma_{\parallel}(\mathbf{r})+\tfrac{1}{3}\sigma_{\perp}(\mathbf{r}) \ . \quad (16)$$

Even this will likely prove too complicated in many composite applications, where the component phases are assumed to be homogeneous. In that case a single conductivity might be assigned to a layer of thickness $\sim\lambda$ at each interface, in effect adding an additional phase to the composite. The conductivity to be assigned would be the volume average of Eq. (16) over this layer.

(iii) The treatment presented here assumes that the active regions for scattering (the "sheaths" surrounding each fiber) do not overlap. This restriction is expressed directly by the limits chosen for the θ integration in Eqs. (7) and (9). Refinements may be possible, but are expected to yield results specific to a particular geometry. Nonetheless, this limitation will have to be overcome if we are ever to describe the transition to low temperatures of metal-matrix composites containing a high density of fibers.

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[1]W. P. Brown, Tran. Soc. Rheol. **9** (1), 357 (1965).

[2]D. Abukay, K. V. Rao, S. Arajs, and Y. D. Yao, Fiber Sci. Tech. **10**, 313 (1977). Data and analysis for other composites in the same series may be found in the doctoral thesis of K. W. Tse, *Electrical Conductivities of Fiber Reinforced Composites* (University Microfilms International, Ann Arbor, MI, 1984), Chap. 4.

[3]R. B. Dingle, Proc. R. Soc. London Ser. A **201**, 545 (1949).

[4]F. S. Roig and J. E. Schoutens, J. Mater. Sci. **21**, 2409 (1986). Subsequently, a correction appeared by the same authors in J. Mater. Sci. **22**, 4002 (1987).

[5]C. A. Moyer, S. Arajs, Y. D. Yao, and Y. Y. Chen, Phys. Status Solidi A **120**, K71 (1990).

[6]R. G. Chambers, Proc. R. Soc. London Ser. A **202**, 378 (1950).

[7]For a more complete discussion of this and related points, see J. M. Ziman, *Electrons and Phonons* (Oxford University Press, Oxford, 1960), Chap. 11.

[8]*Handbook of Mathematical Functions,* edited by M. Abramowitz and I. A. Segun (Dover, New York, 1965), Chap. 5.